

The Electronic States of Two Oppositely doped Mott Insulators Bilayers

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We study the effect of Coulomb interaction between two oppositely doped low-dimensional tJ model systems. We exactly show that, in the one-dimensional case, an arbitrarily weak interaction leads to the formation of charge neutral electron-hole pairs. We then use two different mean-field theories to address the two-dimensional case, where inter-layer excitons also form and condense. We propose that this results in new features which have no analog in single layers, such as the emergence of an insulating spin liquid phase. Our simple bilayer model might have relevance to the physics of doped Mott insulator interfaces and of the new four layer $\text{Ba}_2\text{Ca}_3\text{Cu}_4\text{O}_8$ compound.

In recent years, the advance in material science made it possible to grow atomically smooth epitaxial interfaces between different iso-structural transition metal oxides [1, 2]. In particular, it is possible to grow atomically ordered (doped) Mott insulator interfaces [3, 4]. Knowing the wealth of new physics the interface between (doped) band insulators have produced, and the richer physics of bulk doped Mott insulators as compared to doped band insulators, the interface science of Mott insulators is anticipated to be a promising direction in years to come.

Prompted by the above motivation, in this letter we take the initiative to explore the interfacial electronic states of two low-dimensional doped Mott insulators. We concentrate on the specific case where each Mott insulator is doped with opposite signed charges, and address the possible states caused by the Coulomb attraction between the resulting n and p type carriers. In particular, we find that the formation of electron-hole pairs, also known as excitons, across the interface gives rise to new physics which is absent in the isolated systems.

More specifically, below we study the interface between either two one-dimensional (1D) tJ model chains or two two-dimensional (2D) tJ model layers, where the upper chain (or layer) is doped with a density x of electrons and the lower chain (or layer) has the same density of holes. We first consider the 1D case, where we explicitly show that an arbitrarily small inter-chain Coulomb interaction leads to exciton formation. We then consider the experimentally relevant 2D system which, aside from being an explicit model for engineered (doped) Mott insulator interfaces [3, 4], may also be pertinent to address the recently studied stoichiometric four-layer $\text{Ba}_2\text{Ca}_3\text{Cu}_4\text{O}_8$ compound where the simultaneous presence of n and p type Fermi surfaces indicates the existence of a “self-doping” effect [9]. Unfortunately, the exact argument used in one dimension does not apply in higher dimensions and, in this case, we rather resort two different mean-field schemes, namely, a recently developed fully fermionic mean-field theory [5, 6] and the traditional slave-boson mean-field theory [7, 8]. Although these the-

ories sometimes miss the ultimate low energy physics, they have successfully predicted the qualitative nature of the cuprate phase diagram [5, 8]. Since our current purpose is to explore the possible interface electronic phases, we feel that it is meaningful to work out the prediction of the aforementioned approaches, which we find yield consistent results.

The model. The Hamiltonian describing our interface is $H_{\text{interface}} = \sum_{a=u,d} H_a + H_{\text{int}}$ where

$$H_a = \sum_{\langle ij \rangle \in \text{NN}} J(S_{a,i} \cdot S_{a,j} - \frac{n_{a,i} n_{a,j}}{4}) - t(\tilde{c}_{a,i}^\dagger \tilde{c}_{a,j} + h.c.) \quad (1)$$

is the Hamiltonian for the upper ($a = u$) and lower ($a = l$) systems (chains and layers in the 1D and 2D case, respectively). J and t are the nearest neighbor (NN) antiferromagnetic exchange and electron hopping parameters. $\tilde{c}_{a,i}^\dagger = \mathcal{P}[c_{a,i,\uparrow}^\dagger, c_{a,i,\downarrow}^\dagger]\mathcal{P}$, where $c_{a,i,\sigma}^\dagger$ creates electrons in chain (layer) a . \mathcal{P} projects out doubly occupied (vacant) sites when $a = l$ ($a = u$). $n_{a,i} = \tilde{c}_{a,i}^\dagger \tilde{c}_{a,i}$ and $S_{a,i} = \frac{1}{2} \tilde{c}_{a,i}^\dagger \sigma \tilde{c}_{a,i}$ (σ are the Pauli matrices) are the electron number and spin operators. The across the interface interaction is described by

$$H_{\text{int}} = V \sum_i (1 - n_{u,i})(1 - n_{l,i}) + \frac{\Delta\mu}{2} \sum_i (n_{u,i} - n_{l,i}) \quad (2)$$

where $V > 0$ is the Coulomb repulsion parameter and $\Delta\mu$ is the energy cost to move an electron from the lower to the upper chain (layer).

The exactly solvable 1D interface. We first look at the interface model for two 1D chains. The model then has the remarkable property that in the limit $J \rightarrow 0$ the wavefunction of the ground states factorizes into a spin wavefunction and a charge wavefunction. This interesting feature was first observed in Ref. [10] for the integrable Hubbard chain at large U . However, it is also common to various non-integrable tJ type models, leading to a variety of exact statements [11, 12]. We proceed by performing a particle-hole transformation on the upper chain, which makes doped electrons formally identical to holes, while changing the sign of the interchain

interaction, $V \rightarrow -V$. We then introduce the wavefunction $\psi(x_1, \sigma_1 \dots x_N, \sigma_N; y_1, \chi_1 \dots y_N, \chi_N)$, where x_i, σ_i are the positions and spins of the particles in the upper chain, and y_i, χ_i those of the lower chain. Both sets of coordinates x_i and y_i are assumed to be given in ascending order. The action of the Hamiltonian on the wavefunction ψ now takes the following form

$$H\psi = -t \sum_{l=1}^N (\psi(\dots x_l \pm 1 \sigma_l \dots) + \psi(\dots y_l \pm 1 \chi_l \dots)) - V \sum_{l=1}^N \sum_{m=1}^N \delta_{x_l, y_m} \psi(\dots) + H_J \psi(\dots) \quad (3)$$

where H_J represents the spin exchange term proportional to J . In the limit $J \rightarrow 0$ one can easily verify that the eigenstates of the system described by Eq. (3) are of the following factorized form:

$$\psi(\dots) = f(x_1 \dots x_N; y_1 \dots y_N) g(\sigma_1 \dots \sigma_N; \chi_1 \dots \chi_N). \quad (4)$$

This follows from the fact that at $J = 0$, the Hamiltonian Eq. (3) does not act on the spin degrees of freedom. Hence, f must be an eigenfunction of Eq. (3) with $J = 0$, whereas g can be an *arbitrary* function of spin variables. Precisely at $J = 0$ the ground state is thus hugely degenerate in the spin sector. This degeneracy can be lifted in first order perturbation theory via an effective spin Hamiltonian acting on a spin ladder of $2N$ spins, where N is the number of singly occupied sites in both the upper and the lower chain, and the holes are effectively “squeezed” out of the system [11, 12]. At first order in J , the spin wavefunction g is thus uniquely determined. Here we are, however, more concerned with the charge correlations of the system. In this regard, we observe that the determination of the charge wavefunction f from the Hamiltonian Eq. (3) is equivalent to finding the ground state of the attractive Hubbard model. Indeed, the effective charge Hamiltonian acting on f can be rewritten in second quantized form as

$$H_c = -t \sum_{i,a} (a_{i+1,a}^\dagger a_{i,a} + h.c.) - V \sum_i a_{i,u}^\dagger a_{i,u} a_{i,d}^\dagger a_{i,d} \quad (5)$$

where the fermion operator $a_{i,a}$ carries a “pseudospin” index a labeling the layer. It is well known that Eq. (5) is in the spin gapped Luther-Emery liquid phase of 1D systems for any $V > 0$, which is the 1D analog of a paired superfluid [11]. The spin gap of the Hubbard model hence implies a charge gap due to the formation of electron-hole pairs in the double-chain system. The important lesson to be learned is that, in one dimension, the phase diagram of our interface model features exciton formation at *arbitrarily weak* inter-chain interaction.

The fully fermionic mean-field theory. We now discuss the 2D bilayer Hamiltonian $H_{interface}$ at the mean-field level. We start with the fully fermionic mean-field theory introduced in Refs. 5, 6. This theory introduces fermionic operators $d_{a,i}^\dagger = [d_{a,i,\uparrow}^\dagger d_{a,i,\downarrow}^\dagger]$ that create charge ($+e$) and spin-1/2 holes in the lower Hubbard

band of the lower layer ($a = l$) and charge ($-e$) and spin-1/2 electrons in the upper Hubbard band of the upper layer ($a = u$). The Mott insulating background on top of which these carriers move is described by a singly occupied lattice of neutral spin-1/2 fermions whose creation operators are $f_{a,i}^\dagger = [f_{a,i,\uparrow}^\dagger f_{a,i,\downarrow}^\dagger]$. In terms of the d and f fermions the projected electron operators can be expressed as [6]

$$\begin{aligned} \tilde{c}_{l,i}^\dagger &= s_\sigma \frac{1}{\sqrt{2}} \left[\left(\frac{1}{2} + s_\sigma \tilde{S}_{l,i}^z \right) \tilde{d}_{l,i,-\sigma} - \tilde{S}_{l,i}^{s_\sigma} \tilde{d}_{l,i,\sigma} \right] \\ \tilde{c}_{u,i}^\dagger &= \frac{1}{\sqrt{2}} \left[\left(\frac{1}{2} - s_\sigma \tilde{S}_{u,i}^z \right) \tilde{d}_{u,i,\sigma}^\dagger - \tilde{S}_{u,i}^{s_\sigma} \tilde{d}_{u,i,-\sigma}^\dagger \right] \end{aligned} \quad (6)$$

where $s_\sigma = (+1), (-1)$ for $\sigma = \uparrow, \downarrow$, $\tilde{S}_{a,i} = \frac{1}{2} f_{a,i}^\dagger \sigma f_{a,i}$ and $\tilde{d}_{a,i,\sigma} = d_{a,i,\sigma} (1 - d_{a,i,-\sigma}^\dagger d_{a,i,-\sigma})$. These relations allow us to recast $H_{interface}$ in terms of the d, d^\dagger and f, f^\dagger operators, after what we perform a mean-field factorization to obtain a quadratic Hamiltonian.

First, we summarize the (single layer) mean-field results of Ref. [5, 6]. At low enough doping the tJ model develops antiferromagnetic order [13], which yields non-zero staggered magnetization order parameters

$$\begin{aligned} m &= \frac{1}{2} s_a (-)^{i_x+i_y} \langle \psi_{a,i}^\dagger \psi_{a,i} - 1 \rangle \\ n &= -s_a (-)^{i_x+i_y} \langle \sum_{\nu=2,3} t_\nu \sum_{\hat{u} \in \nu NN} \eta_{a,i}^\dagger \eta_{a,i+\hat{u}} + h.c. \rangle \end{aligned} \quad (7)$$

Here, $\psi_{a,i}^\dagger = [\psi_{a,i,1}^\dagger \psi_{a,i,2}^\dagger] = s_a^{i_x-i_y} [f_{a,i,\uparrow}^\dagger f_{a,i,\downarrow}]$ and $\eta_{a,i}^\dagger = [\eta_{a,i,1}^\dagger \eta_{a,i,2}^\dagger] = s_a^{i_x-i_y} [d_{a,i,\uparrow}^\dagger d_{a,i,\downarrow}]$; $s_a = (+1), (-1)$ for $a = l, u$; $t_2 = J$ and $t_3 = J/2$; $\hat{u} = \pm \hat{x} \pm \hat{y}$ for $\nu = 2$ and $\hat{u} = \pm 2\hat{x}, \pm 2\hat{y}$ for $\nu = 3$ [14]. In addition to the antiferromagnetic order parameter, singlet bond order parameters which capture the spin liquid correlations are also introduced

$$\chi = \langle \psi_{a,i}^\dagger \sigma_z \psi_{a,j} \rangle ; \quad \Delta = (-)^{i_y-j_y} \langle \psi_{a,i}^\dagger \sigma_x \psi_{a,j} \rangle \quad (8)$$

with $\langle ij \rangle \in NN$. Finally, the Bose amplitudes which describe the quantum coherence between the doped carriers and the spin background are given by

$$b_0 = \langle f_{a,i}^\dagger d_{a,i} \rangle ; \quad b_1 = \langle \frac{3}{16} \sum_{\nu=1,2,3} t_\nu \sum_{\hat{u} \in \nu NN} f_{a,i}^\dagger d_{a,i+\hat{u}} \rangle \quad (9)$$

where $t_1 = t$ and $\hat{u} = \pm \hat{x}, \pm \hat{y}$ for $\nu = 1$.

In terms of these order parameters, the in-plane mean-field Hamiltonian is given by [5, 6]

$$\begin{aligned} H_a^{MF} &= \sum_{a,k} \left[\begin{array}{cc} \psi_{a,k}^\dagger & \eta_{a,k}^\dagger \end{array} \right] \left[\begin{array}{cc} \alpha_k^z \sigma_z + \alpha_k^x \sigma_x & \beta_k \sigma_z \\ \beta_k \sigma_z & \gamma_k^z \sigma_z \end{array} \right] \left[\begin{array}{c} \psi_{a,k} \\ \eta_{a,k} \end{array} \right] \\ &+ \sum_{a,k} \left(\nu_a^\psi \psi_{a,k+Q}^\dagger \psi_{a,k} + \nu_{a,k}^\eta \eta_{a,k+Q}^\dagger \eta_{a,k} \right) \end{aligned} \quad (10)$$

In the above equation $\alpha_k^z = -(\frac{3\bar{J}}{4}\chi - t_1 x)(\cos k_x + \cos k_y) + a_0$ where a_0 is the Langrange multiplier that

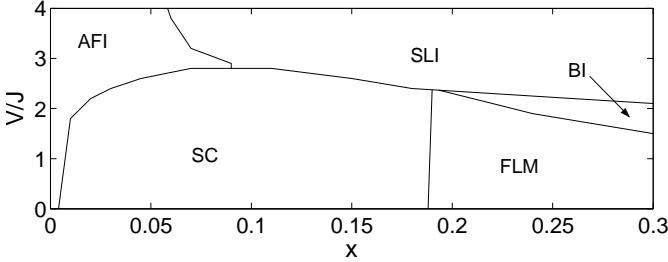


FIG. 1: tJ model bilayer mean-field phase diagram for $t = 3J$. It includes the antiferromagnetic insulator (AFI), the spin liquid insulator (SLI), the superconductor (SC), the band insulator (BI) and the Fermi liquid metal (FLM) described in the main text.

ensures $\langle f_{a,i}^\dagger f_{a,i} \rangle = 1$, and $\tilde{J} = (1-x)^2 J$. In addition, $\alpha_k^x = -\frac{3\tilde{J}}{4}\Delta(\cos k_x - \cos k_y)$, $\beta_k = -\frac{3b_0}{8}[t_1(\cos k_x + \cos k_y) + 2t_2 \cos k_x \cos k_y + t_3(\cos 2k_x + \cos 2k_y)] + b_1$, $\gamma_k^z = t_2 \cos k_x \cos k_y + \frac{t_3}{2}(\cos 2k_x + \cos 2k_y) + \frac{\Delta\mu}{2}$, $\nu_a^\psi = 2s_a(n - 0.34\tilde{J}m)$, $\nu_{a,k}^\eta = 2s_a m (\frac{\Delta\mu}{2} - \gamma_k^z)$.

Unlike other slave-particle approaches [7, 8, 15], all particles in the above mean-field theory are fermions. The central advantage of this mean-field approach is the ease to describe fermionic quasiparticles, which are created by the d^\dagger operator. The “holon” in the slave-boson approach, which is a spin singlet and charge $+e$ non-quasiparticle object, is the composite bosonic particle made up of a d quasiparticle and a f neutral fermion, and its condensation is described by the the order parameters $b_{0,1}$ [6]. The fully fermionic approach, which yields results consistent with numerical calculations [6], successfully reproduces various non-trivial aspects of ARPES and STM experiments in the cuprate superconductors, including the Fermi arcs, the dispersion kink, the minimum gap locus, the flat dispersion around $(\pi, 0)$, the peak-dip-hump and the absence of coherence peaks in deeply underdoped dI/dV spectra [5, 16]. Encouraged by the above successes we extend Eq. (10) to describe two oppositely doped Mott insulator layers. Following our exact 1D result, we introduce the order parameters

$$\begin{aligned} e_s &= \langle \eta_{l,i}^\dagger \sigma_- \eta_{u,i} + \eta_{u,i}^\dagger \sigma_- \eta_{l,i} \rangle \\ e_t &= (-)^{i_x+i_y} \langle \eta_{l,i}^\dagger \sigma_- \eta_{u,i} - \eta_{u,i}^\dagger \sigma_- \eta_{l,i} \rangle, \end{aligned} \quad (11)$$

to describe the formation of spin singlet and spin triplet excitons. In Eq. (11) $\sigma_- = (\sigma_x - i\sigma_y)/2$. The resulting inter-layer mean-field Hamiltonian reads

$$H_{int}^{MF} = -\frac{V}{2} \sum_k \left(e_s \eta_{u,k}^\dagger \sigma_x \eta_{l,k} + i e_t \eta_{u,k+Q}^\dagger \sigma_y \eta_{l,k} + h.c. \right) \quad (12)$$

The zero temperature mean-field phase diagram is determined upon minimizing the mean-field energy $\langle H_u^{MF} + H_l^{MF} + H_{int}^{MF} - E_0 \rangle$, where $E_0 = -[\frac{3\tilde{J}N}{2}(\chi^2 + \Delta^2) +$

$1.36\tilde{J}Nm^2 - 8Nmn - 4Nb_0b_1 + \frac{V}{2}N(e_S^2 + e_T^2) + \Delta\mu N]$ and N is the number of lattice sites. Motivated by the mutual exclusion of antiferromagnetism and superconductivity [4] in the cuprates, we omit the possible coexistence of antiferromagnetic and superconducting orders. The resulting phase diagram for the bilayer $t = 3J$ model is depicted in Fig. 1. It comprises five distinct regions whose properties we discuss below:

(i) *AF insulator* ($\chi, m, n, e_s, e_t \neq 0$ and $b_0, b_1 = 0$). Since $m \neq 0$ spins are antiferromagnetically ordered. Electrically neutral excitons form ($e_s, e_t \neq 0$) which cost a finite energy to break, thus opening a gap in the charge sector.

(ii) *Gapless spin liquid insulator* ($\chi, \Delta, e_s \neq 0$ and $m, n, e_t, b_0, b_1 = 0$). In the absence of AF order ($m = 0$), the spins are in a gapless d -wave ($\Delta \neq 0$) paired spin liquid state reminiscent of the pseudogap state in the single layer problem [8]. However, in the bilayer case excitons ($e_s \neq 0$) render the system a charge insulator.

(iii) *(D + S)-wave superconductor* ($\chi, \Delta, b_0, b_1, e_s \neq 0$ and $m, n, e_t = 0$). Doped carriers hybridize with the d -wave paired spins ($\Delta, b_0 \neq 0$) and, at the same time, participate in forming s -wave excitons. As a result, the superconducting gap acquires a $D + S$ symmetry.

(iv) *Band insulator* ($\chi, b_0, b_1, e_s \neq 0$ and $m, n, e_t, \Delta = 0$). In the absence of pairing ($\Delta = 0$) there is no superconducting order. The non-zero b_0 and e_s condensates open a gap in both the spin and charge sectors and the system behaves as a renormalized band insulator.

(v) *Fermi liquid metal* ($\chi, b_0, b_1 \neq 0$ and $m, n, e_t, e_s, \Delta = 0$). In the absence of Δ and e_s the quasiparticle gap closes and the system behaves as a renormalized Fermi liquid.

The phase diagram in Fig. 1 results from the competition between the spin exchange energy, the kinetic energy (of the doped carriers) and the inter-layer Coulomb energy. The latter two vanish at half-filling, hence, at sufficiently low doping, antiferromagnetic order prevails. As doping increases, the itinerancy of charge carriers tends to destroy antiferromagnetism and to favor a spin liquid state instead [17]. In that case, and for sufficiently large V , the antiferromagnetic insulator is replaced by the spin liquid insulator where all doped carriers form inter-layer excitons. Since charge carriers are more mobile when they are not part of an exciton, partial exciton unbinding is favored at low V . The carriers thus liberated from the excitonic condensate then bind to the spins and, in the presence of spin pairing, the system is a superconductor. At large doping, kinetic energy dominates over the exchange interaction and, as a result, the superconducting gap closes, rendering the bilayer metallic. This metallic state occurs in a system with two electrons per bilayer unit cell and, nominally, it is rather expected to be an insulator. Such band theory expectations are amiss due to the effect of strong correlations [18]. Since the effect of electron-electron correlations decreases upon further

doping, the system eventually becomes a band insulator, as expected in the weak coupling limit.

The slave-boson mean-field theory. To vindicate the above predictions we also determine the phase diagram using the slave-boson mean-field theory [7, 8]. We use the antiferromagnetic (m), in-plane d -wave pairing (Δ^f), inter-plane exciton pairing (Δ^b) and single boson condensation ($\langle b \rangle$) mean-field order parameters and, as before, ignore the coexistence of antiferromagnetic and superconducting orders. To our satisfaction, the results are in qualitative agreement with those obtained using the above fully fermionic mean-field theory. Upon increasing the carrier concentration, the excitonic antiferromagnetic insulator ($m \neq 0$, $\Delta^b \neq 0$, $\Delta^f \neq 0$, $\langle b \rangle = 0$) present at $0 < x < x_c$ evolves to the excitonic d -wave superconductor ($m = 0$, $\Delta^b \neq 0$, $\Delta^f \neq 0$, $\langle b \rangle \neq 0$) for $x > x_c$. The critical concentration x_c increases with V/J in a manner consistent with the AFI-SC phase boundary depicted in Fig. 1. If V/J is larger than a certain value, however, we find that the phase that replaces the AF state is a spin liquid insulator ($m = 0$, $\Delta^f \neq 0$, $\Delta^b \neq 0$, $\langle b \rangle = 0$). A similar AFI-to-SLI transition is found for large V/J values in Fig. 1. Detailed findings of the slave-boson calculation are presented elsewhere [19].

Discussion. Above, we use exact arguments and two different mean-field theories to conclude that Coulomb interaction between oppositely doped tJ model systems leads to the formation of excitons in 1D and 2D, respectively. Since in the 1D [2D] system the hole momentum distribution is shifted from that of electrons by π [(π, π)], excitons carry a finite momentum equal to π [(π, π)] [14].

Even though in the 2D case our model Hamiltonian $H_{interface}$ is, at most, a simplified version of material compounds' Hamiltonians, it provides the playground to explore the important effect of inter-layer Coulomb interaction, which is the largest interaction between neighboring oppositely doped Mott insulators. We also believe that the single layer physics is correctly captured by restricting the in-plane Coulomb interaction to electrons sharing the same lattice site. However, it is well known that terms describing electron hopping beyond NN sites can significantly alter the single plane properties [17, 20]. Above, we neglect such terms, which may limit our ability to address systems like the $Ba_2Ca_3Cu_4O_8$ compound where farther neighbor hopping processes are believed to be important. Although inclusion of longer range hopping is the subject of future work, we expect the present conclusions to apply as long as $xt/t', xV/t' \gtrsim 1$.

Finally, we note that $H_{interface}$ is interesting in itself, as it illustrates how exciting new physics (absent in isolated single layers) may emerge in bilayer systems. In the present case, such physics is summarized as follows:

(i) In the presence of large inter-layer Coulomb interac-

tion all doped carriers form electrically neutral electron-hole pairs and the system is an insulator. As these pairs move throughout the layers, they disorder the spin background and stabilize a fractionalized spin liquid state which supports neutral spin-1/2 excitations. So far, there is no evidence for such spin liquid states in single layers [21], where the itinerant charge carriers render the spin liquid unstable to superconductivity at low temperature. Here, we propose that the charge gap in the excitonic insulating phase can protect the exotic spin liquid state in the bilayer formed by oppositely doped Mott insulators.

(ii) The coexistence of the excitonic condensate and superconductivity changes the quasiparticle gap from pure D to $D+S$ symmetry. The extra gap component deviates the nodes away from the $(0, 0) - (\pi, \pi)$ and $(0, 0) - (-\pi, \pi)$ directions. This effect can, in principle, be detected in ARPES experiments.

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